

Electron effective mass enhancement in Ga(AsBi) alloys probed by cyclotron resonance spectroscopyG. Pettinari,^{1,2,*} O. Drachenko,^{3,†} R. B. Lewis,^{4,5,‡} and T. Tiedje⁴¹*National Research Council, Institute for Photonics and Nanotechnologies (IFN-CNR), Via Cineto Romano 42, 00156, Roma, Italy*²*School of Physics and Astronomy, University of Nottingham, Nottingham NG7 2RD, United Kingdom*³*Dresden High Magnetic Field Laboratory and Institute of Ion-Beam Physics and Materials Research, Forschungszentrum Dresden-Rossendorf, P.O. Box 510119, 01314 Dresden, Germany*⁴*Department of Electrical and Computer Engineering, University of Victoria, Victoria, British Columbia, V8W 2Y2, Canada*⁵*Department of Physics and Astronomy, University of British Columbia, Vancouver, BC V6T 1Z1, Canada*

(Received 27 May 2016; revised manuscript received 15 September 2016; published 19 December 2016)

The effect of Bi incorporation on the conduction band structure of Ga(AsBi) alloys is revealed by a direct estimation of the electron effective mass via cyclotron resonance absorption spectroscopy at THz frequencies in pulsed magnetic fields up to 65 T. A strong enhancement in the electron effective mass with increasing Bi content is reported, with a value of mass $\sim 40\%$ higher than that in GaAs for $\sim 1.7\%$ of Bi. This experimental evidence unambiguously indicates a Bi-induced perturbation of the host conduction band states and calls for a deep revision of the theoretical models describing dilute bismides currently proposed in the literature, the majority of which neglect or exclude that the incorporation of a small percentage of Bi may affect the conduction band states of the host material.

DOI: [10.1103/PhysRevB.94.235204](https://doi.org/10.1103/PhysRevB.94.235204)**I. INTRODUCTION**

Bismuth-alloyed GaAs is a semiconductor system featuring an increasing interest in technological fields ranging from spintronics to long-wavelength devices, from high-efficiency lasers at telecom frequencies to solar cells. Indeed, a few percentage of Bi produces a large bandgap reduction (~ 90 meV/%-of-Bi) and spin-orbit splitting (~ 80 meV/%-of-Bi) with a relatively small change in the lattice constant (6.7×10^{-3} Å/%-of-Bi) [1–3]. Those peculiar and interesting properties of Ga(AsBi) and related materials, such as In(PBi) and Ga(SbBi), are attributed to a resonant interaction between Bi-induced localized states and the valence band states of the host matrix. Such an interaction has been modeled by different theoretical approaches [4–6], which invariably *neglected the role Bi may have on the conduction band of the host material*. Therefore, in Ga(AsBi) only a small monotonic decrease of the electron effective mass is predicted [7,8] as a direct consequence of the narrowing of the bandgap energy with increasing Bi concentration [9]. Recently, two theoretical works [10,11] based on first-principle, *ab initio* calculations have questioned this view and argued that Bi incorporation has an important effect also on the conduction band of the alloy with the electron effective mass that shows a modest monotonic increase with Bi concentration ($1.12 \times 10^{-3} m_0$ /%-of-Bi) [10].

Carrier effective masses are fundamental band structure parameters of a critical importance in materials technology and fundamental research since they rule the system's response to external perturbations. On one hand, they largely determine

the transport and mobility properties of a semiconductor; on the other hand, carrier effective mass measurements have been usefully exploited for probing and disclosing the unusual, puzzling effects that isoelectronic N-doping has in highly mismatched dilute nitride alloys [12–14]. A number of different methods have been proposed in the literature to probe the carrier effective masses in semiconductors, i.e., magnetophotoluminescence spectroscopy, Raman spectroscopy, Seebeck effect measurements, conductivity measurements, etc. However, very often those methods do not give direct access to the effective mass value, the determination of which is subject to the knowledge of other band structure parameters or to data interpretation hypotheses that are not always widely accepted in the research community. In the past, experimental evidence for a Bi-induced effect on the Ga(AsBi) conduction band, reported by some of us by magnetophotoluminescence investigations [15,16], has indirectly suggested an increase in the electron effective mass upon Bi incorporation, although a quantitative estimate of that increase could not be done. Therefore, the effect of Bi on the host conduction states is still an issue strongly debated in the literature [17–20], and a direct determination of the electron effective mass in Ga(AsBi) appears timely and crucial for both fundamental research and technological exploitation of dilute bismide alloys [21].

Cyclotron resonance (CR) absorption spectroscopy is one of the most widely accepted and commonly used techniques to get a direct determination of carrier effective masses. Notwithstanding, this technique is intrinsically challenging and has not been applied so far to dilute bismide alloys since it requires relatively high-quality doped crystals, in order to get a sizable absorption by a high density of free carriers, and/or very high magnetic fields, to satisfy the condition ($\omega_{\text{CR}}\tau \gtrsim 1$) required for a well resolved absorption peak [22,23]. Here, CR absorption spectroscopy at THz frequencies in pulsed magnetic fields up to 65 T has been used to measure the electron effective mass in Ga(AsBi) alloys. The electron effective mass largely increases with increasing Bi content, for $\sim 1.7\%$ of Bi being $\sim 40\%$ higher than that in GaAs. This direct

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evidence points to a *strong perturbation of the host conduction band* upon Bi incorporation, contradicting the widely accepted idea that Bi incorporation does not affect the host conduction band; consequently, theoretical modeling of dilute bismides should be reconsidered and improved.

II. SAMPLES AND EXPERIMENT

We investigated two *n*-type Ga(AsBi) samples grown by molecular beam epitaxy on a GaAs substrate at $T_G = 330$ °C and having Bi concentration (x) of 1.25% and 1.66%, as determined by high-resolution x-ray diffraction experiments. Nominal Si-doping concentrations of 3×10^{18} cm $^{-3}$ and 6×10^{18} cm $^{-3}$ have been supplied during the growth of the two samples, while free-electron concentrations of 3.4×10^{18} cm $^{-3}$ and 7.3×10^{17} cm $^{-3}$ were estimated by room temperature Hall measurements, respectively. The lower free-electron concentration found in the sample with the higher Bi content is a clear indication of a high concentration of compensating acceptors in that sample, likely due to Bi-induced acceptors that have been reported to form in dilute bismides and to have an exponential growth with Bi content [24–26]. A Bi-free GaAs reference sample (Bi-concentration <0.1% as estimated by x-ray diffraction measurements) has also been grown under similar conditions for comparison reasons. Table I reports the main growth parameters along with the relevant physical quantities determined in this paper [27]. Further details on the growth conditions can be found elsewhere [28]. The CR absorption spectroscopy has been performed in the temperature range 4.2–145 K by pulsing the magnetic field up to a value of 65 T, with a pulse cycle of 150 ms, and recording the intensity of a THz monochromatic radiation transmitted through the sample in a Faraday geometry. Quantum cascade lasers (QCLs) at wavelengths of 15 μ m and 66 μ m have been used as generators of THz radiation, which has been detected, respectively, by a Si:B blocked impurity band photodiode or a Ge:Ga extrinsic photoconductor. A detailed description of the experimental setup can be found elsewhere [29].

III. RESULTS AND DISCUSSION

Temperature-dependent CR studies ($T = 4.2$ K, 77 K, and 145 K) have been performed in order to correctly

TABLE I. Summary of properties of the investigated samples. x and t are, respectively, the Bi concentration and the thickness of the Ga(AsBi) layer as determined by high-resolution x-ray diffraction measurements; n_e and μ_e are, respectively, the density and mobility of free electrons, as determined by both direct transport measurements at room temperature and CR spectroscopy; m_e^{CR} is the CR electron effective mass; and τ_e is carrier scattering time as derived within a Drude model [27]. The GaAs reference sample ($x < 0.1\%$) has been grown at low temperature ($T_G = 370$ °C) under conditions similar to those used for Bi-containing samples. Bi surfactant has been used to increase the structural quality of the GaAs reference sample.

Bi content x (%)	Layer thickness t (nm)	From transport measurements		From CR spectroscopy			Scattering time τ_e (fs)
		Electron density n_e (cm $^{-3}$)	Electron mobility μ_e (cm 2 V $^{-1}$ s $^{-1}$)	Electron density n_e (cm $^{-3}$)	Electron mobility μ_e (cm 2 V $^{-1}$ s $^{-1}$)	CR electron effective mass m_e^{CR} (m_0)	
<0.1	667	1.0×10^{17}	3500	$(9.8 \pm 7.1) \times 10^{16}$	2853 ± 60	0.066 ± 0.005	123 ± 23
1.25	300	3.4×10^{18}	927	$(6.8 \pm 0.2) \times 10^{18}$	800 ± 94	0.074 ± 0.001	34 ± 4
1.66	300	7.3×10^{17}	605	$(7.3 \pm 1.2) \times 10^{17}$	588 ± 44	0.095 ± 0.007	44 ± 8

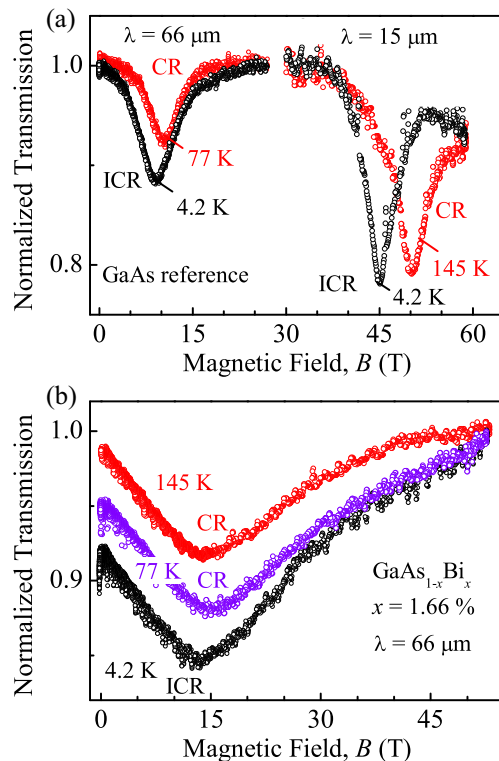


FIG. 1. Normalized transmission spectra recorded (a) on a GaAs reference sample and (b) on a Ga(AsBi) sample with Bi concentration $x = 1.66\%$ for different values of temperature and incident radiation wavelength (indicated in the figure). The CR (red and purple points) and ICR (black point) peaks are identified at different temperatures.

address the effect of Bi incorporation on the conduction band states of the host matrix by disentangling genuine CR free-electron absorption from localized impurity or Bi-cluster related absorptions. Carrier freeze-out on localized states results, indeed, in an impurity-shifted CR (ICR) peak [30], which appears at lower fields than the free-electron CR peak and may dominate the transmission spectra at relatively low temperatures. With increasing temperature, absorption from those ionized states decreases with respect to the free-electron resonant absorption, as shown, for example, in Fig. 1, both in the case of the GaAs reference sample and for the Ga(AsBi)

sample at 1.66% of Bi. In particular, for the Ga(AsBi) sample the CR peak shifts towards high magnetic fields and gets more symmetric by increasing the temperature from 4.2 K to 77 K. A further increase in temperature from 77 K to 145 K does not produce major changes in the CR peak but for a general moderate decrease of the signal, thus indicating that temperatures equal or higher than 77 K are needed to ensure a negligible contribution from ICR absorption to the CR spectrum, while they not distort the CR peak. In the investigated samples, an absorption from Bi-related localized states at $T < 50$ K has been confirmed also by temperature- and power-dependent photoluminescence spectroscopy (not shown here). On this ground, CR experiments have been performed at temperatures high enough to provide a reliable estimate of the effective mass of free electrons in Ga(AsBi).

Figure 2 shows the results of CR experiments on the two Ga(AsBi) samples recorded under condition of temperature and THz radiation optimized independently for each sample to ensure a well-defined CR absorption peak and to reduce the possible influence of absorptions from localized states.

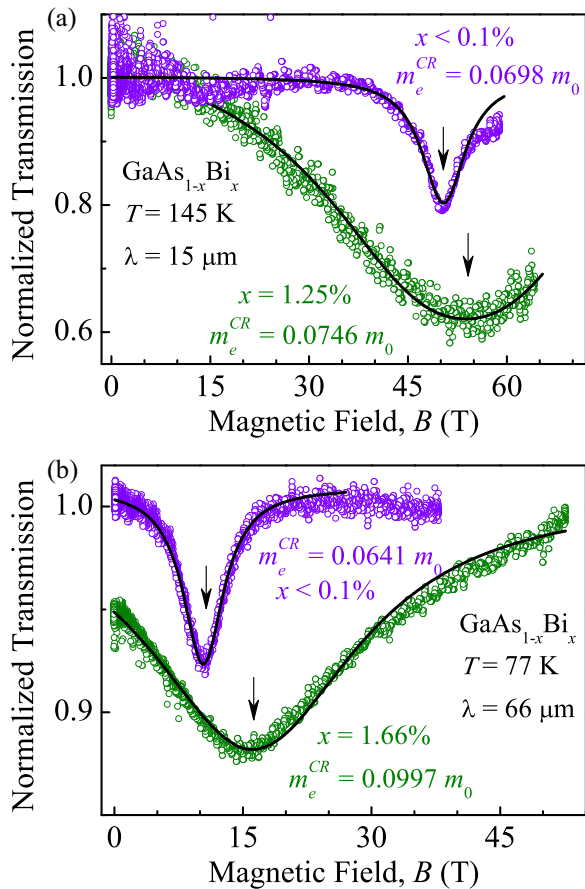


FIG. 2. Normalized transmission spectra recorded on Ga(AsBi) samples (a) with Bi concentration $x = 1.25\%$ and (b) 1.66% . The CR spectra (violet points) recorded on a GaAs reference sample ($x < 0.1\%$) are also shown for ease of comparison. Temperature and incident radiation wavelength are indicated in the figure. Black lines are classical CR absorption fits to the data using Eqs. (2) and (3). Down arrows indicate the minimum of CR spectra (i.e., B_{res}). The best fit values of the electron effective mass (m_e^{CR}) are displayed.

The comparison with CR spectra acquired under the same conditions in the GaAs reference sample clearly indicates a marked shift of the CR peak towards high magnetic fields and a sizable peak broadening in the Ga(AsBi) samples; both of those effects can be easily quantified by classical modeling of CR absorption spectra.

In a semiconductor subject to an external magnetic field (B), a free carrier moves into circular orbits in the plane perpendicular to the field with an angular frequency given by

$$\omega_{\text{CR}} = qB/m^*, \quad (1)$$

where q is the electric charge of the carrier and m^* is its effective mass. Under incident monochromatic electromagnetic radiation and variable magnetic field, as in our experimental configuration, a free carrier can absorb the electromagnetic energy only when the magnetic field is such that ω_{CR} equals the angular frequency of the incident radiation ($2\pi c/\lambda$). Therefore, the magnetic field value ($B_{\text{res}} = 2\pi cm^*/q\lambda$) at which the electromagnetic radiation is resonantly absorbed *depends only on the effective mass of the absorbing carrier*, and thus CR absorption provides a *direct* and easy access to such a fundamental band structure parameter. The lineshape of the CR absorption spectrum provides further information: on the density (n , via the integrated absorption strength of the CR peak) and on the mobility (μ , via the CR spectral linewidth) of the involved free carriers. Indeed, for linearly polarized incident radiation—as that used in our set up—and by considering only one type of absorbing carriers, i.e., electrons or holes, the transmission (T) through the sample is given by the Beer-Lambert law

$$T = A(1 - R)^2 \frac{(e^{-\alpha d} + 1)}{2}, \quad (2)$$

where A is the incident radiation intensity, R and d are the reflectivity and thickness of the absorbing layer, respectively, and α is the absorption coefficient. In turn, α can be expressed as

$$\alpha = \frac{nq\mu/\varepsilon_0 n_1 c}{1 + (2\pi cm^*/q\lambda - B)^2 \mu^2}, \quad (3)$$

where ε_0 is the vacuum permittivity, n_1 is the refractive index of the sample, and λ is the wavelength of the incident radiation. The effective mass determined by this technique is usually called CR *effective mass* (m_e^{CR}) and probes the band structure of the sample around the Fermi level. For n -type doped samples, the CR electron effective mass (m_e^{CR}) is obtained.

The best fit values of the free-electron mass (m_e^{CR}), mobility (μ_e), and density (n_e) obtained by modeling the CR data with Eqs. (2) and (3) are reported in Table I [27]. The values of mobility and electron density extracted by CR spectroscopy well match the values obtained by standard transport Hall measurements at room temperature, thus confirming the correctness of the theoretical model used to fit the CR data and strengthening the reliability of the extracted values of electron effective mass. The good quality of the fitting procedure is also confirmed by good agreement between the value of m_e^{CR} obtained by best fitting the whole CR spectra and the value one can get by simply analyzing the CR peak position in terms of B_{res} .

The values of m_e^{CR} are now compared with the experimental results and theoretical calculations reported in the literature

(see Fig. 3). As mentioned in the Introduction, the majority of the theoretical models describing Bi effects in III-V alloys do not predict direct effects on the conduction band, and the electron effective mass is expected to smoothly decrease (dotted line in the figure; see Ref. [8]) following the narrowing of the bandgap energy with increasing Bi concentration. This widespread view has been questioned first by experimental evidences of a Bi-induced perturbation of the conduction band [15,16] and then by first-principle calculations predicting a moderate increase in the electron effective mass with increasing Bi concentration (dashed line in the figure; see Ref. [10]). On the experimental side, the only values of electron effective mass available so far in the literature are those extracted from the temperature-dependent damping of Shubnikov–de Haas (SdH) oscillations in *n*-type Ga(AsBi) samples for a range of Bi concentrations ($x < 1\%$), quite narrower than that spanned by our paper [17]. Those data, however, do not show a clear compositional trend, as shown by open circles in Fig. 3: The SdH electron effective mass (m_e^{SdH}) first gradually increases with Bi concentration in the range 0%–0.4%, as our data do, then it decreases at higher Bi concentration (0.8%–0.9%), although by an amount comparable with the experimental uncertainty.

On the contrary, the CR measurements we report unambiguously indicate a strong enhancement with Bi concentration of the electron effective mass, with a value of $m_e^{\text{CR}} = (0.095 \pm 0.007)m_0$ for [Bi] = 1.66%, namely a value of mass 40% higher than that in GaAs for a similar doping level. These CR data confirm the early findings of a Bi-induced perturbation of

the host conduction band we have reported by measuring the exciton reduced mass (μ_{exc}) via magneto-optical spectroscopy on intrinsic Ga(AsBi) samples [16,31]. It should be noticed the excellent quantitative agreement between the percentage variation of cyclotron electron effective mass (Δm_e^{CR}) and that of the exciton reduced mass ($\Delta \mu_{\text{exc}}$) with respect to the value in GaAs (see inset in Fig. 3). Such an agreement is expected for an electron effective mass much smaller than the hole effective mass (being $1/\mu_{\text{exc}} = 1/m_e + 1/m_h$), as it is the case of GaAs and of Ga(AsBi) for small Bi content (where a value of $m_h = 0.47 m_0$ has been reported by Landau-level spectroscopy) [32]. In this respect, it is worth recalling that the magnetophotoluminescence spectroscopy on intrinsic Ga(AsBi) samples probes the band structure at the Γ point (i.e., at $k = 0$) [16], while the CR spectroscopy on doped samples probes the band structure around the Fermi level (i.e., at $k_F = 5.9 \times 10^8 \text{ m}^{-1}$ and $2.8 \times 10^8 \text{ m}^{-1}$ for the 1.25% and 1.66% sample, respectively). Notwithstanding, the two sets of data match each other, thus suggesting on one side limited or negligible nonparabolic effects on the conduction band of Ga(AsBi) alloys, at least up to $\sim 1.7\%$ of Bi. A parabolic conduction band in Ga(AsBi) is suggested also by the value of the electron effective mass ($0.074 m_0$) found in the 1.25% sample for a doping level of $\sim 7 \times 10^{18} \text{ cm}^{-3}$. Such a value, indeed, is sizably lower than that reported for a similar doping level in GaAs ($\sim 0.082 m_0$) as a result of the conduction band nonparabolicity [33]. Finally, the results reported in the inset of Fig. 3 confirm that the magnetophotoluminescence spectroscopy can be a valid and complementary technique to directly address the transport properties even for highly disordered semiconductor systems, as demonstrated here for the case of dilute bismides.

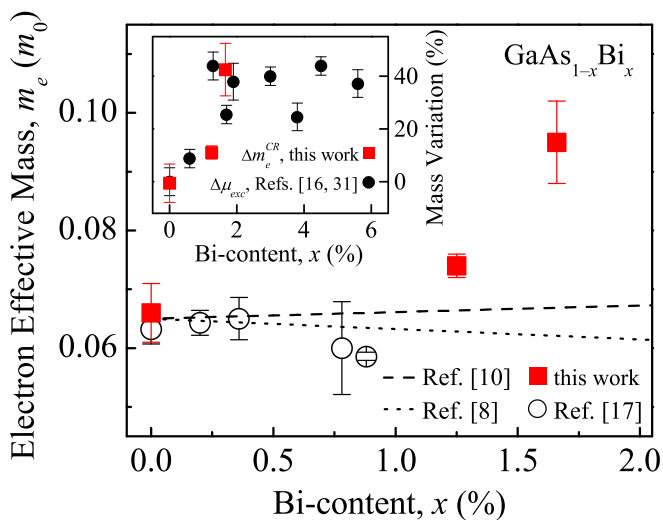


FIG. 3. Compositional dependence of the electron effective mass in Ga(AsBi). The experimental values of CR effective mass (m_e^{CR} , red squares) are compared with values deduced by SdH measurements (m_e^{SdH} , open circles; see Ref. [17]) and theoretical expectations based on the valence band anticrossing model (dotted line; see Ref. [8]) or first-principles calculation (dashed line; see Ref. [10]). Inset: Variation of different effective masses in Ga(AsBi) with respect to the value in GaAs as a function of Bi content [$\Delta f(x) = f(x)/f(x=0) - 1$]. The variation of the CR electron effective mass (Δm_e^{CR} , red squares) are compared with that of the exciton reduced mass as deduced by the magnetophotoluminescence spectroscopy on similar samples ($\Delta \mu_{\text{exc}}$, black circles; see Refs. [16] and [31]).

IV. CONCLUSIONS

In summary, a direct experimental evidence of a strong electron effective mass enhancement in Ga(AsBi) alloys upon Bi incorporation has been reported, with a value of effective mass 40% higher than that in GaAs for [Bi] $\sim 1.7\%$. These results, obtained by CR absorption spectroscopy at THz frequencies and high magnetic fields, confirm early indirect evidences of Bi-induced perturbations on the host conduction band [15,16,19] and call for a deep revision of the theoretical models currently proposed in the literature. The majority of those models, indeed, neglect the effects that the incorporation of a small percent of Bi atoms may have on the conduction band states of the host materials. Moreover, our result opens interesting scenarios in which Bi alloying can be exploited to engineer the conduction as well as the valence band of III-V semiconductors and will drive further investigations and innovative applications of the peculiar electronic properties of dilute bismides.

ACKNOWLEDGMENTS

Part of this work has been supported by the European Union (under Grant No. PIEF-GA-2010-272612), by Italian Ministry of Education, Universities and Research (under Futuro in Ricerca di Base project DeLIGHTeD, Prot. RBFR12RS1W),

by German Deutsche Forschungsgemeinschaft (German Research Foundation) (Grant No. DR832/3-1), and by EuroMag-NET II (under European Council Contract No. 228043). G.P.

acknowledges A. Patanè (The University of Nottingham) for fostering this work and A. Polimeni and M. Capizzi (Sapienza University of Rome) for useful discussions.

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